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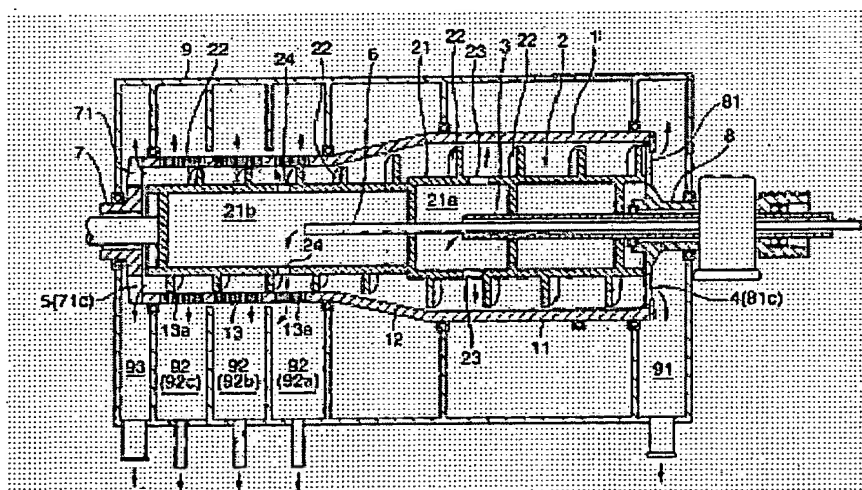
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(54) Title: METHOD FOR MANUFACTURING AROMATIC DICARBOXYLIC ACID



(57) Abstract: A method for producing an aromatic dicarboxylic acid involving oxidizing an aromatic compound having an alkyl or partially oxidized alkyl substituent by a gas containing molecular oxygen in a reaction solvent in the presence of a catalyst and then: subjecting the resultant slurry containing crystals of the aromatic dicarboxylic acid to a soil-liquid separation, to recover the crystals, wherein the slurry is continuously fed to a screen type centrifuge having a screw conveyor arranged therein to carry out the soil-liquid separation, characterized in that a screen having an aperture through which a part of crystals in the slurry leak is used as the screen for the screen type centrifuge. The method allows the prevention of the clogging of a screen type centrifuge, which leads to the achievement of an efficient soil-liquid separation.



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(57) Abstract (Translation of abstract in source document):

A method for manufacturing an aromatic dicarboxylic acid in which an aromatic compound having an alkyl substituent or a partially oxidized alkyl substituent is subjected to liquid-phase oxidation with a molecular oxygen-containing gas in a reaction solvent in the presence of a catalyst, a slurry that contains crystals of the resulting aromatic dicarboxylic acid is subsequently subjected to solid-liquid separation, and the crystals are recovered, with the method for manufacturing an aromatic dicarboxylic acid being characterized in that when solid-liquid separation is performed by continuously supplying the slurry to a screen-type centrifugal separator with an internally disposed screw conveyor, a screen whose opening size allows the crystals in the supplied slurry to partially pass therethrough is used as the screen of the screen-type centrifugal separator. According to the method, clogging of the centrifugal separator can be prevented and solid-liquid separation can be performed in an efficient manner.

DESCRIPTION

METHOD FOR MANUFACTURING AROMATIC DICARBOXYLIC ACID

TECHNICAL FIELD

The present invention relates to a method for manufacturing an aromatic dicarboxylic acid, and more particularly to a method for manufacturing an aromatic dicarboxylic acid using a screen-type centrifugal separator with an internally disposed screw conveyor when subjecting a slurry containing crystals of an aromatic dicarboxylic acid obtained from a reaction to solid-liquid separation.

BACKGROUND ART

Aromatic dicarboxylic acids are manufactured by means of a method comprising subjecting an aromatic compound having an alkyl substituent or a partially oxidized alkyl substituent to liquid-phase oxidation with a molecular oxygen-containing gas in a reaction solvent in the presence of a catalyst, subjecting the slurry that contains crystals of the resulting aromatic dicarboxylic acid to solid-liquid separation, and recovering the crystals.

Techniques using a screen-type centrifugal separator with an internally disposed screw conveyor have been proposed (e.g. WO 98/18750) as solid-liquid separation methods, as described in the foregoing.

When a screen is used to perform solid-liquid separation, it is important for the screen to be prevented from getting clogged so that separation can be performed efficiently. Clogging is especially exacerbated in screen-type centrifugal separators with screw conveyors by layers of solidified crystals (cakes) that form in a compacted state on the inside of the screen.

JP (Kokai) 8-294643 proposes a scraper for scraping off the solidified crystal layers in the screen to maintain its function, but a problem is presented insofar as the system becomes complicated due to the presence of the scraper. Furthermore, as relates to the size of the screen openings, it is stated in e.g. "Filtration & Separation (Sep., 2000, p. 245)" that the size of the screen openings should preferably be set to be two to three times the particle size of the solid to

be separated in order to prevent the particles from becoming lodged in the screen and clogging it. Nevertheless, such large-mesh screens have the problem of allowing too much of the material to pass through the screen openings.

With the foregoing aspects in view, it is an object of the present invention to provide a method for manufacturing an aromatic dicarboxylic acid that has been improved as a result of using a screen-type centrifugal separator with an internally disposed screw conveyor to enable clogging of the centrifugal separator to be prevented and solid-liquid separation to be conducted in an efficient manner when subjecting slurries containing crystals of an aromatic dicarboxylic acid obtained from a reaction to solid-liquid separation. It is another object of the present invention to provide a method for manufacturing an aromatic dicarboxylic acid that has been improved as a result of having the crystals that are recovered by means of solid-liquid separation exhibit an increased level of purity.

DISCLOSURE OF THE INVENTION

In order to achieve the above objectives, the present inventors conducted diligent investigations into the layers of solidified crystals (cakes) formed in a compacted state by the screw on the inside of the screen, and obtained the findings hereunder as a result. In other words, if particles of indeterminate form and relatively small size in the slurry are allowed to pass through the screen openings, then layers of solidified crystals (cakes) that form in a compacted state on the inside of the screen from particles of indeterminate form and relatively large size will not cause significant clogging to occur. Moreover, when the particles of indeterminate form are crystals, impurities will be concentrated in those crystals having a relatively small particle size; therefore, when such crystals pass through the openings as described above, the purity of the recovered crystals will increase.

The present invention has been perfected based of the above findings. The main point of the present invention lies in a method for manufacturing an aromatic dicarboxylic acid in which an aromatic compound having an alkyl substituent or a partially oxidized alkyl substituent is subjected to liquid-phase oxidation with a molecular oxygen-containing gas in a reaction solvent in the presence of a catalyst, a slurry that contains crystals of the resulting aromatic dicarboxylic acid is subsequently subjected to solid-liquid separation, and the crystals are recovered, with the

method for manufacturing an aromatic dicarboxylic acid being characterized in that when solid-liquid separation is performed by continuously supplying the slurry to a screen-type centrifugal separator with an internally disposed screw conveyor, a screen whose opening size allows the crystals in the supplied slurry to partially pass therethrough is used as the screen of the screen-type centrifugal separator.

The present invention is described in detail hereunder.

First, the liquid-phase oxidation reaction shall be described. In the present invention, a slurry containing an aromatic dicarboxylic acid is obtained as a result of subjecting an aromatic compound having an alkyl substituent or a partially oxidized alkyl substituent to liquid-phase oxidation with a molecular oxygen-containing gas in a reaction solvent in the presence of a catalyst.

An aromatic compound having an alkyl substituent or a partially oxidized alkyl substituent is used as the starting material for the reaction. Monocyclic or polycyclic aromatic compounds may be used. Examples of alkyl substituents include C₁ to C₄ alkyl groups such as methyl, ethyl, *n*-propyl, and isopropyl groups, while examples of partially oxidized alkyl groups include aldehyde, acyl, carboxyl, and hydroxyalkyl groups.

Specific examples of aromatic compounds having an alkyl substituent (alkyl-substituted aromatic hydrocarbons) include dialkyl benzenes such as *m*-diisopropylbenzene, *p*-diisopropylbenzene, *m*-cymene, *p*-cymene, *m*-xylene, and *p*-xylene; dialkyl naphthalenes such as dimethylnaphthalenes, diethylnaphthalenes, and isopropylnaphthalenes; and dialkyl biphenyls such as dimethyl biphenyls.

Specific examples of aromatic compounds having a partially oxidized alkyl substituent include 3-methylbenzaldehyde, 4-methylbenzaldehyde, *m*-toluic acid, *p*-toluic acid, 3-formylbenzoic acid, 4-formylbenzoic acid, and formyl naphthalene carboxylic acids.

Combinations of heavy metal compounds and bromine compounds can be used as the catalyst. Examples of heavy metals in heavy metal compounds include cobalt, manganese, nickel, chromium, zirconium, copper, lead, hafnium, and cerium. Examples of compounds of heavy metals include acetates, nitrates, acetylacetates, naphthenates, stearates, and bromides.

Examples of bromine compounds include inorganic bromine compounds such as molecular bromine, hydrogen bromide, sodium bromide, potassium bromide, cobalt bromide, and manganese bromide; and organobromine compounds such as methyl bromide, methylene bromide, bromoform, benzyl bromide, bromomethyl toluene, dibromoethane, tribromoethane, and tetrabromoethane.

The ratio of the bromine compound to the heavy metal compound is selected from a range of usually 0.05 to 10 moles, and preferably 0.1 to 2 moles, as expressed in terms of the ratio of bromine atoms per mole of heavy metal atoms. The amount of the catalyst to be used is selected from a range of usually 10 to 10000 ppm, and preferably 100 to 5000 ppm, as expressed in terms of the concentration of heavy metal in the reaction solvent.

A lower aliphatic carboxylic acid is preferably used as a reaction solvent. Specific examples of such lower aliphatic carboxylic acids include acetic acid, propionic acid, and butyric acid. The lower aliphatic carboxylic acids can be used alone or as a mixture with water. The amount of the reaction solvent to be used is selected from a range of usually 0.5 to 70 parts by weight, and preferably 2 to 50 parts by weight, as expressed in terms of the ratio per part by weight of the starting material in the liquid-phase region.

Examples of the molecular oxygen-containing gas include oxygen and air. The molecular oxygen-containing gas is supplied in an excess of the quantity necessary for oxidizing the aromatic compound as a starting material into an aromatic carboxylic acid. When air is used, the amount thereof to be used is selected from a range of usually 2 to 20 Nm³, and preferably from 2.5 to 15 Nm³, as expressed in terms of the ratio per kilogram of aromatic compound used as a starting material for oxidation.

The reaction is ordinarily carried out at normal temperature, and the reaction time is selected from a range of usually 4 to 180 minutes, and preferably 6 to 120 minutes. When the oxidation reaction is carried out continuously, the reaction time is the residence time. A part of the aromatic dicarboxylic acid produced as a result of the reaction precipitates as crystals, and another part dissolves in the solvent to form a slurry. For instance, when terephthalic acid is manufactured by means of subjecting *p*-xylene to liquid-phase oxidation, the slurry is usually composed of acetic acid as a reaction solvent, terephthalic acid crystals, a catalyst dissolved in

the reaction solvent, unreacted starting material, by-products, unprecipitated terephthalic acid, and the like. The average particle size of the terephthalic acid crystals is usually $120\ \mu\text{m} \pm 40\ \mu\text{m}$.

Solid-liquid separation shall be described hereunder. In the present invention, the slurry containing the crystals of the aromatic dicarboxylic acid obtained from the above reaction is subjected to solid-liquid separation, and the crystals are recovered. The crude crystals recovered from the reaction step are therefore usually treated in a purifying step, and a high-purity aromatic dicarboxylic acid is recovered as a result of subjecting the slurry formed in the purification step to solid-liquid separation. The crystals can also be washed during solid-liquid separation. Lower aliphatic acids such as acetic acid are favorably used as the cleaning fluid.

The solid-liquid separation is carried out using a screen-type centrifugal separator with an internally disposed screw conveyor. In the present invention, it is possible to use screen-type centrifugal separators having a variety of structures, regardless of their descriptive name, as long as solid-liquid separation can be performed using a screen via the action of a centrifugal force while the material to be treated is transported with a screw conveyor. Examples known in the art include a "decanter-type centrifugal separator" (JP (Kokai) 7-155643, et al.) and a "screen-bowl decanter centrifugal separator" (JP (Kokai) 2000-350946, WO 98/18750, et al.).

The screen-type centrifugal separator shown in FIG. 1 has an outer rotating cylinder (1), a screw conveyor (2) that is mounted inside the outer rotating cylinder while allowed to rotate in relative fashion and that comprises a cylindrical rotating shaft (21) and a screw (22), and a slurry supply pipe (3) that is disposed within the rotating shaft of the screw conveyor and that supplies a slurry to the interior of the shaft. The proximal end side of the screw conveyor (2) is provided with a slurry supply port (23) for supplying the slurry to the outer rotating cylinder (1). The outer rotating cylinder (1) is sequentially constituted from a large-diameter region (11) on the proximal end side, a slanted region (12) of a gradually decreasing diameter, and a small-diameter region (13) on which a screen (13a) is provided. An overflow port (4) is provided at the proximal end of the large-diameter region (11), and a solids discharge port (5) is provided at the distal end of the small-diameter region (13).

In a preferred embodiment, the interior of the rotating shaft (21) of the screw conveyor is partitioned into a slurry supply region (21a) on the proximal end side and a cleaning fluid supply region (21b) on the distal end side. The slurry supply region (21a) is designed to be capable of supplying the slurry via the slurry supply pipe (3), and the cleaning fluid supply region (21b) is designed to be capable of supplying the cleaning fluid via a cleaning fluid supply pipe (6) inserted into the interior of the slurry supply region (3). A cleaning fluid supply port (24) through which cleaning fluid is supplied to the small-diameter region (13) of the outer rotating cylinder (1) is provided on the distal end side of the screw conveyor (2).

A plurality of openings (71c) are provided to the outer peripheral region of a flange (71) of a drive shaft (7) for the outer rotating cylinder, and these openings constitute the solids discharge port (5). Openings (81c) are provided to the outer peripheral region of a flange (81) of a drive shaft (8) for the screw conveyor to constitute the overflow port (4).

All of these elements are housed in a casing (9). The inside of the casing (9) is divided with the help of partitioning walls into an overflow liquid reservoir (91), a filtrate reservoir (92), and a solids reservoir (93); and the filtrate reservoir (92) is further divided with the help of partitioning walls into three compartments (92a), (92b), and (92c) along the longitudinal direction of the small-diameter region (13) for the convenience of filtrate sampling. Discharge piping is provided respectively for the overflow liquid reservoir (91); the compartments (92a), (92b), and (92c) of the filtrate reservoir (92); and the solids reservoir (93).

An outstanding feature of the present invention is that a screen with an opening size that allows a portion of the crystals in the supplied slurry to pass therethrough is used as the screen of the screen-type centrifugal separator. The phrase "a portion of the crystals ... to pass therethrough" means that crystals in an amount equivalent to 1 wt% or more will pass through the screen openings.

In other words, the opening size of the screen used in the present invention must be of a magnitude that enables the crystals to pass continuously in an amount equivalent to 0.01 A to 0.1 A (kg/hr), with A (kg/hr) being the rate of the crystals in the continuously supplied slurry. The passage of crystals through the screen openings is believed to occur preferentially with those

of small particle diameters; in the present invention, therefore, crystals having relatively small diameters in the slurry are allowed to pass through the screen openings.

The opening size is defined in detail hereunder. In other words, when the average particle size of the crystals in the slurry is B (μm), the opening size of the screen is usually no less than $(B - 10)$ μm and no more than $(B + 80)$ μm , while preferably being no less than $(B - 5)$ μm and no more than $(B + 60)$ μm . Consequently, when the average particle size of the crystals is 100 μm , the opening size of the screen will usually be 90 to 180 μm , and preferably 95 to 160 μm .

The opening size generally represents the minimum width of the holes, and can be determined according to the shape of the holes in the screen in the following manner. In other words, the opening size refers to the width when the holes are slit-shaped, to the diameter when the holes are circular, to the minor diameter when the holes are elliptical, to the length of the shorter side when the holes are rectangular, to the length of one side when the holes are square, to the shortest distance between the two parallel sides when the holes are rhomboid, and to the shortest distance between the longer sides when the holes are parallelogramic in other configurations.

The particle size of the crystals in the slurry can be measured with the help of a variety of well-known methods used to measure the particle sizes of powdered materials, but the values measured with the help of a laser-scattering particle size distribution measuring device have been adopted in the present invention.

Consequently, with the present invention, the solidified layers of the crystals (cake layers) formed in a compacted state in the inside of the screen, and specifically in the spaces between the outer rotating cylinder (1) and the screw (22), are constituted from crystals with relatively large particle sizes. The reason that excessive clogging does not occur with such solidified crystal (cake) layers has yet to be definitively clarified, but the explanation is presumed to be as provided hereunder.

In other words, when particles of indeterminate form and relatively small size are contained in the solidified crystal (cake) layers, such cakes typically become firm and solid due

to a strong bridging action caused by the presence of said particles. On the other hand, solidified crystal (cake) layers composed of particles of indeterminate form and of relatively large size tend to collapse readily due to the absence of a strong bridging action. Consequently, the solidified crystal (cake) layers in the present invention are presumed to continuously collapse and rearrange, and are accordingly not believed to cause excessive screen clogging.

The clogging preventive effect cannot be achieved with screens having an opening size that causes the pass rate to be less than 1 wt%. On the other hand, with screens having too large an opening size, the anticlogging effect may reach saturation while the pass rate becomes too high, thereby resulting in an inefficient operation. Consequently, the pass rate is preferably no higher than 40 wt%, more preferably no higher than 20 wt%, and even more preferably no higher than 10 wt%.

Moreover, impurities are concentrated in crystals of relatively small particle size, and their passage elevates the purity of the recovered crystals. In other words, when terephthalic acid is manufactured as a result of subjecting *p*-xylene to liquid-phase oxidation, the recovered crystals will have a low concentration of *p*-toluic acid, which is a typical impurity.

In the present invention, solid-liquid separation is performed in the following manner. A slurry is supplied to the large-diameter region (11) from the slurry supply pipe (3) via the slurry supply region (21a). The slurry is then subjected to solid-liquid separation as a result of the centrifugally separating action of the outer cylinder (1) that rotates at high speed.

The separated liquid is taken out from the overflow port (4) via the overflow liquid reservoir (91). Meanwhile, the crystals are transferred from the slanted region (12) to the small-diameter region (13) with the help of the screw (22). The mother liquor in the crystals is also separated by way of the centrifugal force through the screen (13a). The cleaning fluid is simultaneously supplied from the cleaning fluid supply pipe (6). The cleaning fluid is sprayed on the moving crystals through the cleaning fluid supply port (24) via the cleaning fluid supply region (21b). The cleaned and dehydrated crystals are extracted from the solids discharge port (5) via the solids reservoir (93).

There are no particular limitations as to the conditions employed for the solid-liquid separation, but it is preferable for a centrifugal force of 300 to 5000 G to be exerted on the screen and for the residence time of solid-liquid separation on the screen to be set at 2 to 20 seconds. The residence time is a condition relating to the time after the slurry has passed the cleaning fluid supply zone in cases where a cleaning fluid supply port has been provided.

If the centrifugal force is less than 300 G, the residence time for solid-liquid separation on the screen will be too high, and when the centrifugal force exceeds 5000 G, the centrifugal separator will become difficult to operate in a stable manner. The preferred range of centrifugal force is 500 to 3000 G. The centrifugal force is adjusted by controlling the number of rotations of the motor that drives the outer rotating cylinder (1).

If the residence time is less than two seconds, solid-liquid separation will be impossible to perform to a sufficient degree, while if the residence time exceeds 20 seconds, the solid-liquid separating effect will reach saturation, resulting in the centrifugal separator operating at reduced efficiency. The preferred residence time is three to five seconds. The residence time can be adjusted either by means of a method whereby an appropriate design is selected for the size (length) of the screen, or as a result of controlling the operating conditions whereby an appropriate difference in rotational speed is selected for the outer rotating cylinder (1) and the screw conveyor (2) (i.e., the transfer speed of the treated material across the screen).

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a cross-sectional illustrative diagram of a screen-type centrifugal separator that is suited for use in the present invention.

BEST MODE FOR CARRYING OUT THE INVENTION

The present invention is described in detail below with reference to working examples; however, the present invention shall not be construed to be limited to these examples, and may be embodied in other forms within the purview of the invention.

Working Examples 1 and 2 and Comparative Example 1

A slurry containing crystals of terephthalic acid (solids concentration: 30 wt%; average particle size: 100 μm ; *p*-toluic acid (P-TA) concentration in the solid: 180 ppm) obtained from a terephthalic acid manufacturing step was supplied continuously at 40°C under normal pressure to a screen-type centrifugal separator shown in FIG. 1 having screens (of a slit type) with the opening sizes shown in Table 1, with the slurry being subjected to solid-liquid separation under the conditions shown in Table 1. The centrifugal force values in Table 1 refer to the values on the screen. The results are shown in Table 1.

The average particle size of the terephthalic acid crystals was a reading of a particle size that had a cumulative value of 50% and was based on a particle size distribution measured using a particle size distribution meter ("LASER MICRON SIZER LMS-24") manufactured by Seishin Enterprise Co., Ltd. The screen opening size was determined as the average value by magnifying the slit portions using a microscope and measuring the width of the openings in 360 locations.

Working Example 3

Solid-liquid separation was conducted in the same manner as described in Working Example 1, except for the use of a slurry containing terephthalic acid crystals with an average particle size of 110 μm and a solids concentration of 30 wt%, a screen (slit type) with an opening size of 150 μm , and conditions of 160 °C and 0.65 MPa. The results are shown in Table 1.

Table 1

	Screen opening size (μm)	Amount of slurry treated (t/hr)	Centrifugal force (G)	Ratio of crystals passing through screen openings (wt%)	Average particle size of crystals that passed through screen openings (μm)	Recovered crystals	
						Liquid content (wt%)	P-TA concentration (ppm)
Working Ex. 1	100	1.1	750	3.0	60	11	—
Working Ex. 2	100	1.5	1000	5.2	64	10	157
Comparative Ex. 1	70	1.1	750	0.5	15	20	—
Working Ex. 3	150	6.0	750	5.9	110	11	—

As is evident from the results shown in Table 1, the liquid content of the solids recovered in Working Examples 1 to 3 was lower than in Comparative Example 1. This is attributable to the fact that clogging was prevented, which allowed the liquid to be satisfactory removed.

Reference Examples 1 to 6

In order to confirm the effect of the residence time, solid-liquid separation was conducted in the same manner as described in Working Example 1 as a result of employing the conditions shown in Table 2 (with the centrifugal force on the screen being 750 G). The liquid content of the recovered cakes is shown in Table 2.

Table 2

	Residence time (sec)	Slurry supply rate (kg/hr)	Liquid content of recovered cakes (wt%)
Reference Ex. 1	3.2	1110	13.0
Reference Ex. 2	3.2	1480	12.7
Reference Ex. 3	5.6	1110	11.6
Reference Ex. 4	5.6	1480	11.1
Reference Ex. 5	0.8	1110	19.4
Reference Ex. 6	0.8	1480	21.4

The residence time shown in Table 2 refers to the time after the slurry has passed through the cleaning fluid supply zone where the cleaning fluid is sprayed from the supply port (24) (and more specifically refers to the time after the point at which two seconds has elapsed after the slurry has passed directly under the cleaning fluid supply port (24)). The residence times and the liquid content of the recovered cakes were determined in the following manner for the sake of convenience. In other words, the filtrate specimens were recovered from the positions corresponding to the particular residence times shown above, measurements were made of the concentration of solids in the filtrate specimens for each residence time, and the liquid content of the recovered cakes was determined by way of material balance calculations.

The results shown in Table 2 clearly indicate that in Reference Examples 1 to 4, the residence times were appropriate, which allowed the centrifugal separator to be operated efficiently and cakes having a sufficiently reduced liquid content to be recovered. With Referential Examples 5 and 6, on the other hand, the recovered cakes had a high liquid content due to residence times being short.

INDUSTRIAL APPLICABILITY

According to the present invention as described in the foregoing, clogging of the centrifugal separator is prevented when a slurry containing crystals of an aromatic dicarboxylic acid obtained from a reaction is subjected to solid-liquid separation. Also, according to the present invention, the purity of crystals is enhanced. Therefore, the present invention is of high commercial value.

CLAIMS

1. A method for manufacturing an aromatic dicarboxylic acid in which an aromatic compound having an alkyl substituent or a partially oxidized alkyl substituent is subjected to liquid-phase oxidation with a molecular oxygen-containing gas in a reaction solvent in the presence of a catalyst, a slurry that contains crystals of the resulting aromatic dicarboxylic acid is subsequently subjected to solid-liquid separation, and the crystals are recovered, with the method for manufacturing an aromatic dicarboxylic acid being characterized in that when solid-liquid separation is performed by continuously supplying the slurry to a screen-type centrifugal separator with an internally disposed screw conveyor, a screen whose opening size allows the crystals in the supplied slurry to partially pass therethrough is used as the screen of the screen-type centrifugal separator.

2. The manufacturing method according to Claim 1, wherein a screen with an opening size that allows the passage of an amount of crystals equivalent to 1 to 10 wt% in the supplied slurry is used as the screen of the screen-type centrifugal separator.

3. The manufacturing method according to Claim 1 that uses a screen-type centrifugal separator, wherein the screen-type centrifugal separator has an outer rotating cylinder (1), a screw conveyor (2) is mounted inside the outer rotating cylinder while allowed to rotate in relative fashion and that comprises a cylindrical rotating shaft (21) and a screw (22), and a slurry supply pipe (3) that is disposed within the rotating shaft of the screw conveyor and that supplies a slurry to the interior of the shaft; the proximal end side of the screw conveyor (2) is provided with a slurry supply port (23) for supplying the slurry to the outer rotating cylinder (1); the outer rotating cylinder (1) is sequentially constituted from a large-diameter region (11) on the proximal end side, a slanted region (12) of gradually decreasing diameter, and a small-diameter region (13) on which a screen (13a) is provided; an overflow port (4) is provided at the proximal end of the large-diameter region (11); and a solids discharge port (5) is provided at the distal end of the small-diameter region (13).

4. The manufacturing method according to Claim 3, wherein an interior of the rotating shaft (21) of the screw conveyor is partitioned into a slurry supply region (21a) on the

proximal end side and a cleaning fluid supply region (21b) on the distal end side; the slurry supply region (21a) is designed to be capable of supplying the slurry via the slurry supply pipe (3), and the cleaning fluid supply region (21b) is designed to be capable of supplying the cleaning fluid via a cleaning fluid supply pipe (6) inserted into the interior of the slurry supply region (3); and a cleaning fluid supply port (24) through which cleaning fluid is supplied to the small-diameter region (13) of the outer rotating cylinder (1) is provided on the distal end side of the screw conveyor (2).

5. The manufacturing method according to Claim 1, wherein the residence time for solid-liquid separation on the screen is set at 2 to 20 seconds, provided that when a cleaning fluid supply port is provided, a condition is established whereby the residence time is counted after the passage of the cleaning fluid supply zone, and a centrifugal force of 300 to 5000 G is exerted on the screen.

6. The manufacturing method according to Claim 1, wherein the average particle size of crystals in the slurry is 80 to 160 μm .

7. The manufacturing method according to Claim 6, wherein when the average particle size of crystals in the slurry is B (μm), the opening size of the screen is no less than $(B - 10) \mu\text{m}$ and no more than $(B + 80) \mu\text{m}$.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP02/08606

A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl.⁷ C07C51/265, 51/43, 51/47, 63/26, B04B3/04

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho	1922-1996	Jitsuyo Shinan Toroku Koho	1996-2002
Kokai Jitsuyo Shinan Koho	1971-2002	Toroku Jitsuyo Shinan Koho	1994-2002

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	WO 98/18750 A1 (Mitsui Chemicals, Ltd.), 07 May, 1998 (07.05.98), Claims; drawings & EP 889021 A1 & US 5925786 A	1-7
Y	JP 10-36313 A (Tsukishima Kikai Co., Ltd.), 10 February, 1998 (10.02.98), Claims; drawings (Family: none)	1-7
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☐ Further documents are listed in the continuation of Box C.
 ☐ See patent family annex.

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Date of the actual completion of the international search
07 November, 2002 (07.11.02)Date of mailing of the international search report
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